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Positronium annihilation in high-purity liquids

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ABSTRACT

The positron lifetime measurements have been performed as a function of temperature for liquid methane and propane and as a function of magnetic field for liquid argon. The long o-Ps lifetime, high o-Ps intensity, and a strong temperature dependence of these values are observed, and interpreted as being due to the Ps localization in the bubble state. The bubble size and surface tension are evaluated using bubble model and o-Ps lifetime. The chemical reaction of Ps atom with oxygen impurities was studied. The reaction rate K_{ps} and its temperature behavior in liquid methane and propane were obtained. The results of magnetic field experiments in both pure and oxygen-added liquid argon show that the lifetime of o-Ps with magnetic quantum number m=0 state decreases basically according to the normal Zeeman effect.

I. Introduction

In recent years, there has been a growing interest in the study of the behavior of Positronium (Ps), the bound state of a positron and an electron, in liquid system[1,2]. The Ps atom is found to be localized in a microscopic cavity, called a Ps bubble state. The bubble model was first suggested by Ferrell[3] to explain an unexpectedly long o-Ps lifetime in liquid helium. The bubble radius was determined by a balance between the outward pressure caused by the Ps zero-point motion and the shrinking forces due to surface tension and external pressure. In the presence of a chemically reactive species, the o-Ps lifetime is significantly reduced by a process called chemical quenching.The strength of chemical quenching depends on the chemical and physical properties of the molecules. The application of static magnetic field causes a mixing of p-Ps and the o-Ps with m=0 states and changes their annihilation parameters. Along this line of work, the liquid Ar is a special system. In absence of magnetic field, the p-Ps intensity measured by ACAR is not equal to 1/3 of o-Ps intensity existing reported by positron lifetime experiments[1]. This discrepancy remains unresolved for many years. For magnetic quenching experiments, the ACAR results show an unexpected small increase of the narrow component[4]. In this paper, we report the results of positron lifetime measurements in high purity liquid CH_4, C_3H_8 and Ar to study the Ps bubble state, chemical reaction, and magnetic quenching.

II. Experiments

The research grade gas (CH_4, C_3H_8 or Ar) was first passed through a purifier to reduce oxygen and other impurities to less than 1 ppm and then condensed to solid (CH_4 and C_3H_8) or liquid (Ar) in a sample cell made of aluminum which was mounted on a cool head of closed-cycle He refrigerator with a temperature control better than ± 0.1K over a range from 10 to 300 K. For oxygen quenching experiments, the oxygen and pure gas were first mixed in the gas line and then condensed . A ^{22}Na positron source, which sealed between two thin Al foils (6μm) was used in the liquid experiments. The positron lifetime measurements were performed using conventional fast-fast coincident system. For magnetic quenching, the scintillators and magnetically shield photomultiplier were separated by lucite light pipes (18 inch) in order to eliminate the effect of magnetic field. The time resolution of the system was 280 ps for temperature experiments and 400 ps for magnetic quenching.

We measured the positron lifetime spectra as a function of temperature for liquid CH_4 and C_3H_8, and as a function of magnetic field ranging from 0 to 2.0 T for liquid Ar. The lifetime spectra were analyzed using the PATFIT computer program[5]. In absence of magnetic field, three lifetime components were resolved, and the τ_1,τ_2,τ_3 are assigned to p-Ps, positron and o-Ps annihilation ,respectively. For liquid Ar, in the presence of magnetic field, the spectra were analyzed by four components by constraining the longest lifetime $\tau_4=\tau_3$ of liquid Ar at zero field due to the annihilation of o-Ps with m=±1 and $I_3/I_4=1/2$ according to the population of ground states of o-Ps. A source correction was made in data analysis.

III. Results and Discussions

1. Ps-bubble State

Positron lifetime measurement has been provided a wealth of useful information concerning bubble formation in high purity liquid. A sharp transition from solid to liquid was observed[6], i.e., from 2.3 to 4.8 ns, 1.2 to 2.9 ns and 2.4 to 5.0 ns for high purity liquid CH_4, C_3H_8 and Ar, respectively. The results indicated that the annihilation sites of Ps in liquid are different from that in solid. The Ps is believed to localized in bubble state which is a result of strong exchange repulsion between the electron of Ps and the tightly bound electron in the medium.

The temperature dependencies of o-Ps lifetime and intensity of liquid CH_4 and C_3H_8 were shown by solid cycles in Fig.1-2, and Fig.3-4, respectively. From Fig.1 and Fig.3, the o-Ps lifetimes increase with the increasing temperature, i.e. from 4.8 to 6.4 ns for CH_4 and from 2.9 to 3.8 ns for C_3H_8. However, the variations of o-Ps intensities shown in Fig.2 and Fig.4 have different trend, i.e. while the temperature increased, the o-Ps intensity of liquid CH_4 is nearly constant, but a slight increase of o-Ps intensity was observed for liquid C_3H_8. The reason may be the different physical properties in liquids CH_4 and C_3H_8. According to the Ps bubble model and measured o-Ps lifetime, the surface tension and bubble size in liquid CH_4 are evaluated and shown in Table 1 along with other physical parameters[6].

Finally, we have measured positron lifetime spectra in liquid Ar at 86 K and solid Ar at 60 K. In solid Ar, we observed a I_3 of 8.77% with a τ_3=2.38 ns, which agree with the existing reported results [7,8]. However, in liquid Ar, we observed a large value of I_3=24.48%, which agrees with the estimated value of I_3=22 ± 4% by ACAR measurement[1], while the existing lifetime results show only in the order of 10%. The large difference of o-Ps intensity between the present work and the existing results is thought due to the presence of impurity in the Ar sample.

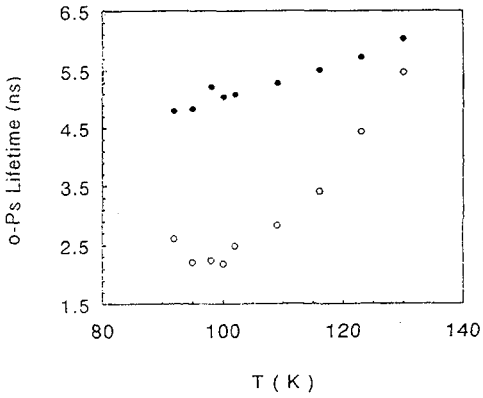

Fig.1 The variations of o-Ps lifetime with temperature in pure (●) and O_2-added (o) liquid CH_4

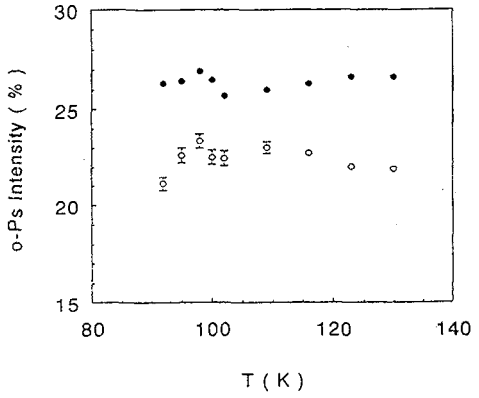

Fig.2 The variations of o-Ps intensity with temperature in pure (●) and O_2-added (o) liquid CH_4

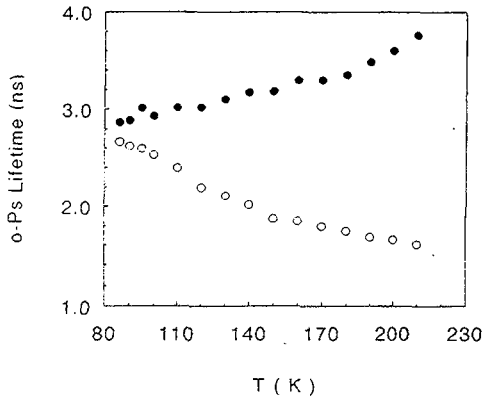

Fig.3 The variations of o-Ps lifetime with temperature in pure (●) and O_2-added (o) liquid C_3H_8

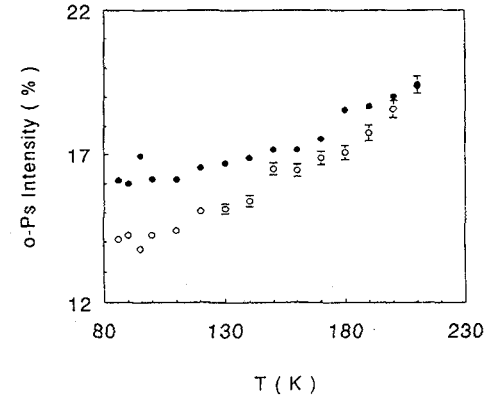

Fig.4 The variations of o-Ps intensity with temperature in pure (●) and O_2-added (o) liquid C_3H_8

Table 1. The bubble radius (R) and surface tension (γ) for liquid methane

T(K)	τ_3(ns)	Density (gcm^{-3})	Pressure (Torr)	R(Å)	γ(dyn cm^{-1})
93	4.85 ± 0.04	0.4499	350	4.67	18.0
95	4.94 ± 0.04	0.4472	465	4.71	17.6
102	5.09 ± 0.04	0.4378	517	4.78	16.3
109	5.29 ± 0.04	0.4280	724	4.87	15.1
116	5.57 ± 0.04	0.4180	1134	5.00	13.8
123	5.73 ± 0.03	0.4077	1758	5.07	12.5
131	6.04 ± 0.04	0.3951	2895	5.19	11.0
140	6.56 ± 0.04	0.3797	4705	5.40	9.4
150	7.28 ± 0.04	0.3611	7497	5.67	7.5

2.Chemical Reaction with Oxygen

Oxygen is known as an effective chemical quencher. We measured the positron lifetime spectra as a function of temperature for liquid CH_4 and C_3H_8 containing 6.875×10^{-2} Mol and 6.0×10^{-2} Mol oxygen, respectively. The results of o-Ps annihilation parameters are also shown in Fig.1-4 by the cycles. We found that the effects of oxygen quenching on o-Ps annihilation parameter are quite different for liquid CH_4 and C_3H_8. Furthermore, the reaction rate K_{ps} can be evaluated using the following equation[2]

$$\lambda_3=\lambda^0_3+K_{ps}[M] \quad (1)$$

where [M] is the molar concentration of oxygen, and λ^0_3 and λ_3 are o-Ps annihilation rates in pure and in oxygen-added liquid respectively. One of the most fascinating observations in Ps chemistry is the variation of K_{ps} with temperature[9]. We plot the $\ln(K_{ps})$ against 1/T in Fig.5-6 for liquid CH_4 and C_3H_8. For liquid CH_4, the K_{ps} increases with temperature at low temperature, and reaches a maximum value at T_0 (100K), the so-called turnover temperature, then decreases above T > 100K. A normal Arrhenius behavior at T < 100K and opposite that at T > 100K were observed in Fig.5. So the "normal" and "negative" activation energies E_a=3.70 KJ/Mol and E'_a=-8.68 KJ/Mol were obtained from least square fitting of the right and left portions of the curve. For liquid C_3H_8, in the temperature range of 85-210 K, we only observed the increase of K_{ps} with the increasing temperature, and the normal activation energy E_a=3.28 KJ/Mol was evaluated.

3. Magnetic Quenching

Theoretical aspects of magnetic quenching of Ps annihilation have been developed by Halpern and by Mills [10,11]. The application of magnetic field has no effect on positron nor on o-Ps with m= ± 1 states, but has an effect on p-Ps and o-Ps with m=0 states. The annihilation rates λ_1(H) and λ_3(H) for the mixed p-Ps and o-Ps with m=0 states as a function of external magnetic field H follow[12]:

$$\lambda_1(H)=(\lambda_1(0)+a^2\lambda_3(0))/(1+a^2) \quad (2)$$

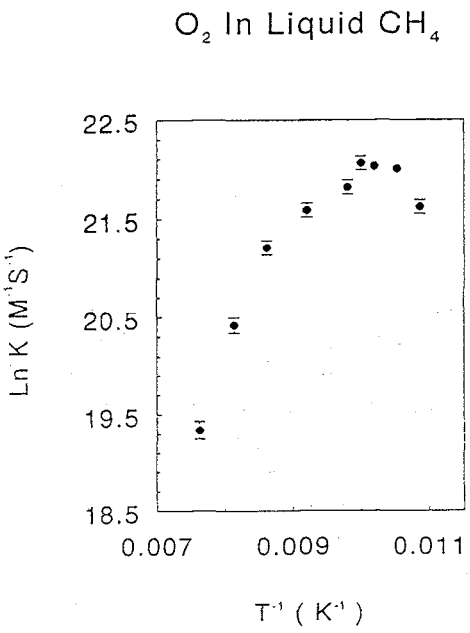

Fig.5 The plot of $\ln(K_{ps})$ vs. 1/T in chemical reaction for Ps with oxygen in liquid CH_4

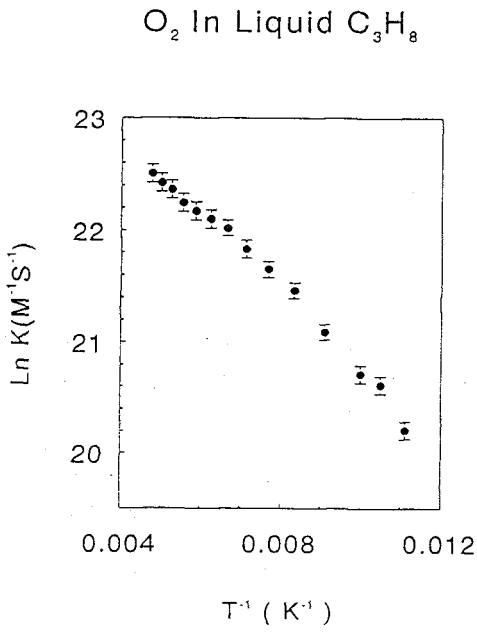

Fig.6 The plot of $\ln(K_{ps})$ vs. 1/T in chemical reaction for Ps with oxygen in liquid C_3H_8

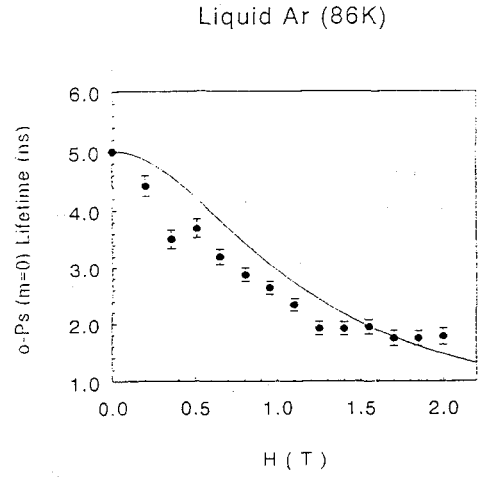

Fig.7 The lifetime of o-Ps with m=0 in liquid Ar at 86K as a function of magnetic field. The solid line is theoretical value for η=1

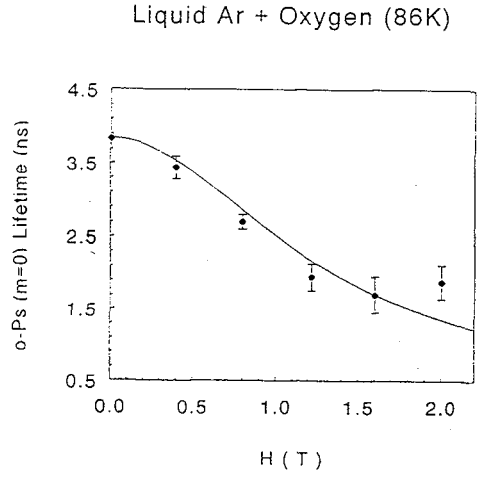

Fig.8 The lifetime of o-Ps with m=0 in O_2-added liquid Ar at 86K as a function of magnetic field. The solid line is theoretical value for η=1

$$\lambda_3(H)=(\lambda_3(0)+a^2\lambda_1(0))/(1+a^2) \quad (3)$$

with

$$a=(\sqrt{1+x^2}-1)/x \quad (4)$$

$$x=4\mu_B H/\Delta E \quad (5)$$

$$\Delta E=\eta\Delta E_{vac} \quad (6)$$

where μ_B is the Bohr magneton, η is used to describe the effect of medium on the Ps wavefunction and very close 1 for the Ps trapped in a bubble state, ΔE is the hyperfine splitting and $\Delta E_{vac}(=8.45\times10^{-4}eV)$ is the vacuum value of ΔE. Consequently, the measured lifetime spectra contain two distinct o-Ps components contributed from o-Ps with m=0, and from o-Ps with m= ± 1 states, respectively. The lifetime of o-Ps with m=0 decreases as a function of magnetic field while the lifetime with m= ± 1 states is unchanged. In four-lifetime analysis, they are designated as τ_3 and τ_4, for m=0 and m= ± 1, respectively. The τ_1 increases slightly but τ_3 decreases largely with magnetic field according to eqs.(2) and (3), respectively. Since τ_1 only slightly increases from 0.122 to 0.130 ns for the magnetic field ranging from 0 to 2.0 T. So we analyzed all lifetime spectra with a fixed value of τ_1=0.125 ns. The results of τ_3, the lifetime of o-Ps with m=0 state, as a function of magnetic field are plotted in Fig.7. As expected, τ_3 decreases with increasing of the field. The variation of τ_3 basically follows the theoretical value plotted by solid line for η=1. As expected we observe nearly constant values of τ_2 and I_2. The sum of o-Ps intensities (i.e. I_3+I_4) is also nearly constant value indicates that the magnetic field has no effect on the Ps formation mechanism, at least up to 2.0 T. In oxygen-added liquid Ar, the lifetimes τ_3 of o-Ps with m=0 state vs H are shown in Fig.8 as a function of magnetic field. The variation of τ_3 follows the theoretical prediction for η=1. These results indicate the lifetime of o-Ps with m=0 state decreases basically according to the normal Zeeman effect.

REFERENCES

1. Stewart,A.T., Briscoe,C.V., and Steinbacher,J.J., Can. J. Phys., 68(1990)1362
2. Schrader,D.M. and Jean,Y.C., eds., "Positron and Positroniun Chemistry" (Elsevier, Amsterdam, 1988)
3. Ferrell,R.A., Phys. Rev., 108(1957)167
4. Steinbacher,J.J. and Stewart,A.T., Mat. Sci. Forum, 105-110(1992)1711
5. Kirkegaard,P., Eldrup,M., Mogensen,O.E. and Pedersen,N.J., Comp. Phys. Comm., 23, (1981)307
6. Wang,S.J., Nakanishi,H. and Jean,Y.C., J. Phys.:Condens. Matter, 3,(1991)2167
7. Jean, Y.C. Yu, C., Zhou, D.M. Phys. Rev. B., 32 (1985) 4313
8. Rytsola, K., Vettenranta, J., Hautojarvi,P., J.Phys. B. 17 (1984) 3359
9. Nakanishi,H., Wang,S.J. and Jean,Y.C.,in "Positron Annihilation", edited by L.Dorikens-Vanpraet et al.(World Sci. Singapore,1988),p740
10. Halpern,O., Phys. Rev., 94(1954)904
11. Mills, Jr., A.P., J.Chem. Phys. 62 (1975) 2647
12. Dupasquier,A., De Natale,P. and A. Rolando, Phys. Rev. A, 43(1991)10036